

Atmospheric Pollutants and Their Effect on Riming Within Orographic Clouds

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ABSTRACT

Presented is an in depth look at past research regarding the relationship between pollutants and riming within clouds. Past research conducted by Borys et al. (2003) demonstrates that an increase in Cloud Condensation Nuclei (CCN), which is indicative of increased pollution, is correlated with a decrease in cloud droplet diameter, and a decrease in riming. A study was conducted at the Storm Peak Laboratory (SPL) in Steamboat Springs, CO, during the week of March 11-18, 2006, validating the work of Borys et al. (2003). The 2006 data from SPL shows that as CCN concentrations increase, cloud droplet diameter and riming levels decrease. According to Borys et al. (2003) and Lohmann (2004), changes in riming levels may also affect subsequent precipitation values or snow water equivalents which affect water levels in the Western United States, but further research is necessary to determine a relationship.

INTRODUCTION

Riming in clouds has been the subject of numerous studies over the past several decades. Riming is the process by which ice crystals “collide with supercooled droplets, which freeze on contact and stick together” (Ahrens, 1991). Supercooled droplets are droplets that remain in a liquid phase even though temperatures are at or below freezing (0°C). Riming plays a role in cloud physics and the process is directly affected by numerous atmospheric constituents. Atmospheric pollution has been on the rise

since the dawn of the Industrial Revolution. Increased pollutants signify an increase in the number of possible Cloud Condensation Nuclei (CCN) in the atmosphere due to an increase in atmospheric aerosols, or particulate matter which is suspended in the air. According to Borys et al. (2003), as CCN concentration increases within a cloud, the average size of cloud droplets tends to decrease. A decrease in cloud droplet diameter can decrease the riming efficiency and therefore total riming within clouds. The current research project performed on the week of March 11-18, 2006, at the Storm

Peak Laboratory in Steamboat Springs, CO, demonstrates the relationships between CCN concentrations, cloud droplet diameter, and riming levels on snow crystals, thereby validating the work of Borys et al. (2003).

Winter orographic clouds, or clouds developing due to processes along mountain ranges, are typically made up of millions of liquid droplets and ice crystals (Hindman et al., 1983). Many of these liquid cloud droplets exist in a supercooled state. In order for this water to freeze, microscopic impurities “with a crystalline structure similar to ice,” called ice nuclei, must be present in the water (Raubert et al., 2002). In a cloud, these ice nuclei are referred to as cloud condensation nuclei (CCN). Common examples of CCN include dust, organic particles, ocean salt, chemical particles released from anthropogenic sources, etc. CCN are not present in all of the droplets that make up a cloud, and many types of CCN are not activated until temperatures cool below -10°C to -15°C ; hence, clouds with tops that are warmer than -15°C , which is typical of many winter orographic clouds, will tend to have supercooled droplets (Raubert et al., 2002).

CCN are related to riming efficiency. CCN are directly proportional to the number of droplets within a cloud. As CCN and the number of cloud droplets increase, the average size of the cloud droplets tends to decrease (Borys et al., 2003). The vapor within the cloud becomes more dispersed amongst the increased CCN, thus droplets decrease in size. “The riming efficiency rapidly approaches zero for all snow crystal sizes if the cloud droplet size decreases below $10\mu\text{m}$ in diameter” (Lohmann, 2004; Pruppacher and Klett, 1997). Thus, if a cloud has a high number of CCN, the cloud droplet number will increase, while the droplet size will decrease, thereby decreasing the riming efficiency within the cloud. Riming efficiency is decreased

because the cloud droplets may not become large enough to reach a terminal velocity and fall through the cloud. Even if particular droplets or ice crystals do become large enough to fall through the cloud due prolonged riming processes, the other cloud droplets may be too small to be used efficiently in the collision coalescence process as the droplet or ice crystal falls through the cloud. If cloud droplets and supercooled droplets are too small, they can be swept around falling hydrometeors (rain droplets or snow crystals) rather than riming or coalescing, and thus riming or coalescence efficiency is decreased.

Much previous work has been conducted surrounding the topic of rime ice and related variables over the past two decades leading researchers to more current theories. Hindman et al. (1983) examined the riming rates versus the precipitation rates in the Colorado Rockies, and found that riming rates were seven times that of the snowfall precipitation rates, showing that riming is an important atmospheric process meriting further research. Berg et al. (1990) concluded that high values of conductivity for snow and low pH values for snow would indicate more riming on snow crystals. Thus, factors affecting conductivity and pH values of cloud droplets, such as atmospheric pollution, may affect riming levels. In 1986 through 1987, the chemical composition of rime ice and pure snowfall was analyzed at five different orographic sites in the state of California, with locations including sites in the central Sierra Nevada range, and also sites located downwind of Los Angeles. Upon completion of this intensive observation period (IOP), Berg et al. (1991) concluded that chemical concentration of the rime ice was significantly larger than the chemical concentration of the snow at all of the sites. The report suggests that, “rime is a source of concentrated chemical constituents,” and

further research should be conducted to test this theory. Hindman et al. (1994) examined the physical and chemical properties of cloud droplets over ten winters (1983/84-1992/93) atop the mountains in Colorado. The study concluded that clouds containing droplets with more acidic pH values were the clouds with the largest cloud droplet concentrations and the smallest average cloud droplet diameters. These studies suggest that atmospheric pollutants, which can affect chemical composition and pH levels of rime, could possibly affect the CCN concentration and cloud droplet diameter and thereby influence riming efficiency.

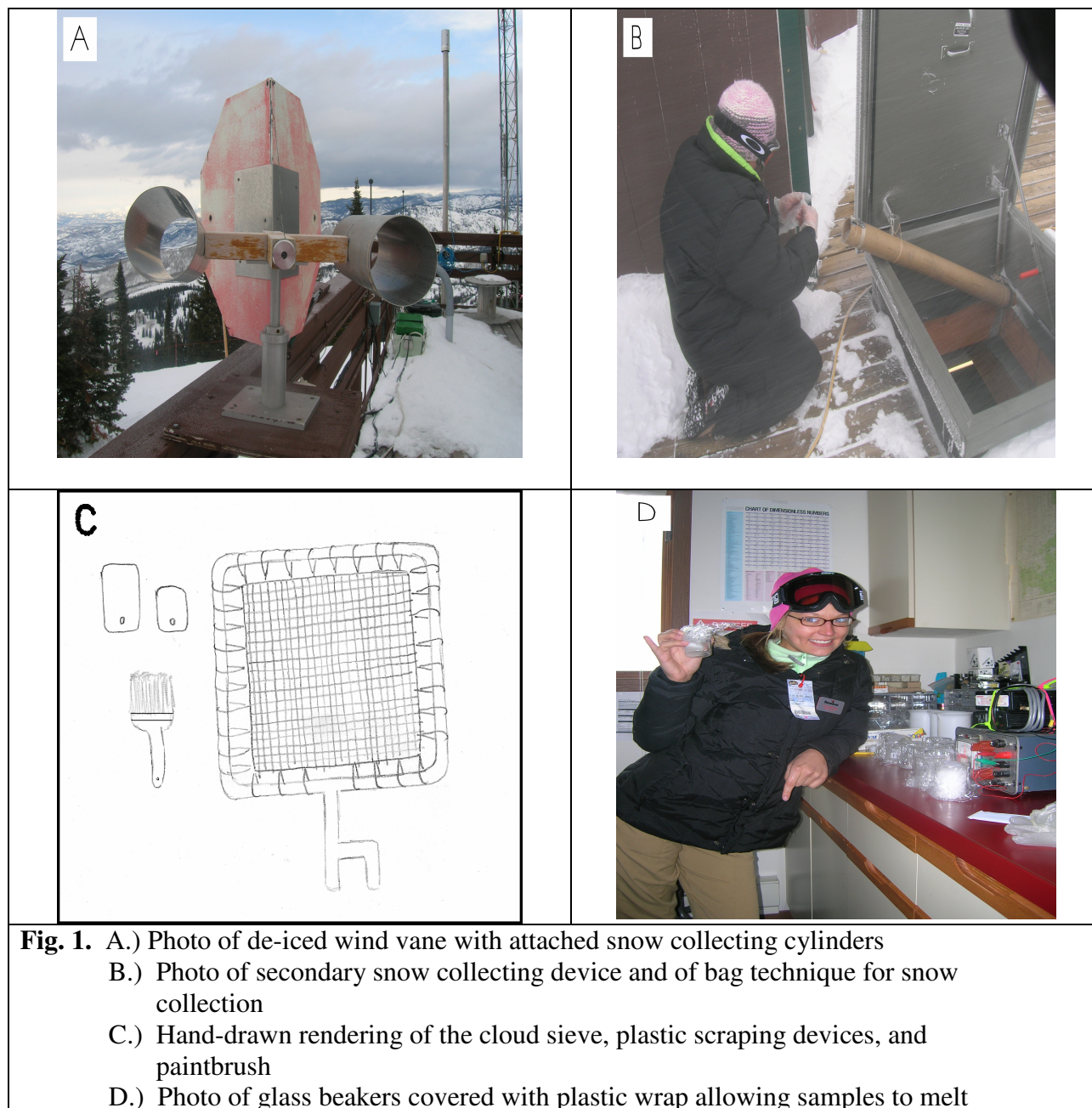
These previous topics lead the scientific community to more current research issues. Borys et al. (1999) concluded that increased concentrations of CCN due to anthropogenic sources led to a decrease in cloud droplet size thus somewhat inhibiting the riming process. This conclusion was reinforced again by Borys et al. in 2003. The 2003 study compared two specific snow events; a rimed case and an unrimed case. Researchers compared the clear air equivalent anthropogenic sulfate concentrations between the two cases. During the rimed event the sulfate concentrations were 8.9 times lower than those of the unrimed case. This shows that more anthropogenic CCN may lead to less riming due to decreased droplet sizes. The research within this project seeks to verify the current findings of Borys et al. (1999, 2003), or to corroborate the second indirect aerosol effect. The second indirect aerosol effect refers to an increase in CCN leading to an increase in the number of cloud droplets, but a decrease in the diameter of the droplets. The researchers expect that as polluted air masses move into the Colorado and specifically Steamboat Springs region during the week of March 11-18, 2006, the CCN concentrations will increase. This

increase is expected to lead to a decrease in cloud droplet diameter and thus a decrease in riming onto snow crystals. The validation of this basic theory lends credibility to further claims proposed by Borys et al. (2003).

Borys et al. (2003) also showed that decreasing riming levels were correlated with lower snow water equivalent precipitation rates. Snow water equivalent refers to the amount of water in a melted unit of snow. This was refuted by Lohmann (2004), who stated that riming levels and snowfall rates also depend on the size and shape of the crystal, and this was neglected by Borys et al. (2003). Thus Lohmann (2004) claims that a decrease in riming does not necessarily point to a decrease in snow water equivalent precipitation rates. (The above mentioned studies will be addressed further in the Discussion section of this paper.) The implications of these studies become very pertinent when considering the lack of water in the Western United States. Water rights are considered a valuable property in the Western States, because control of water resources can be highly lucrative as the water is divided amongst domestic, industrial, and agricultural uses. The Colorado River is considered “the lifeblood of the West,” and this river is fed by snow melt off of the Rocky Mountains (Tripoli, 2006). If pollution is decreasing riming in the clouds along the Rocky Mountains and possibly decreasing the snow water equivalent precipitation rates in these areas, this could lead to a prolonged drought in the Western United States.

METHODOLOGY

To test the second indirect aerosol effect, snow and rime ice were collected within 15-19 minute periods during two separate snow events at the Storm Peak Lab. Researchers were aided by expert Dr. Randolph Borys, in



equipment usage, and experimental technique. Samples were collected on the roof of the Storm Peak Lab facility in Steamboat Springs, CO, at 10,525 ft, atop Mt. Werner (see Fig. 1 for examples of equipment and measurement techniques). Before each collection period, a DMT-SPP 100 Droplet Sizing Probe was turned on to collect cloud droplet data, including CCN concentration and cloud droplet

diameter, during the sample collection period. Snow and rime ice samples were collected on the roof of the Storm Peak Laboratory. Time was recorded at the beginning of the collection period. Wind direction was determined by looking at a de-iced wind vane. A cloud sieve used for size-specific capture of supercooled droplets was then placed in slots so that the face of the cloud

sieve was perpendicular to the wind. This wind is then passing through the cloud sieve allowing the cloud droplets to rime onto the intertwined wires. Three wind measurements were then taken using the hand-held wind measurement gage. During all collection periods, neoprene gloves were worn to prevent contamination of the samples. The plastic bags were then placed inside two hollow cylinders attached to wind vane. The bags were pulled out slightly and folded over the edges of the cylinders, and were rubber-banded on the outside of the cylinders. These cylinders were then adjusted and positioned to catch snow in the blowing wind. At one point during collection, the winds were too strong to use the hollow cylinders attached to the wind vane. The bags were rubber-banded to the cylinders, but the strong winds blew the bags out of the back of each cylinder. A secondary snow-collection device was created. A thick hollow cardboard cylinder was attached to the hatch entrance on the roof of the lab. The cylinder was placed at a lower level, so the winds would not destroy the snow measurements. The plastic bags were again placed inside the cylinder, were folded over the outer edge of the cylinder, and were rubber-banded in place. The cardboard cylinder was placed so that it was parallel to the wind and was left out for the duration of the collection period.

Before taking the roof equipment down, three more wind measurements were recorded. A small black board covered in felt was also held into the wind in order to collect snow crystals for microscopic examination of the snow crystals collected during the period. The cloud sieve was then removed from its slots, and the plastic bags were pulled out of the snow-collecting tubes. The equipment was then taken, very carefully, down from the roof, and into the cold room. Samples were then collected.

In the cold room, the cloud sieve was placed over a heavy-duty garbage bag, which was laid flat on a counter. Plastic scraping devices were then used to scrape the rime off of the cloud sieve onto the garbage bag taking care that nothing tainted the sample. A soft-bristled paint brush was then used to sweep the rime sample into a plastic bag. The snow and rime samples were then weighed using the scale, and the mass was recorded. The board covered with black felt and snow crystals from the event was then placed under the microscope, and snow crystals were assessed qualitatively for riming levels. Classification techniques followed the five degree system proposed by Mitchell and Lamb (1989). The degree of riming will be “classified into five categories according to the fraction of the crystal surface covered by accreted drops: ‘light or 1’ (less than about 1/5), ‘light-to-moderate or 2’ (1/5 to 2/5), ‘moderate or 3’ (2/5 to 3/5), ‘moderate-to-heavy or 4’ (3/5 to 4/5), and ‘heavy or 5’ (greater than 4/5, including graupel)” (Mitchell and Lamb, 1989).

Samples were then removed carefully from the collection bags and were placed in glass beakers. The beakers were sealed with plastic wrap to avoid evaporation, and samples were allowed to melt and reach room temperature. While samples melted, a conductivity meter, and pH probe were calibrated. The conductivity and pH levels of the melted rime ice and snow were then measured, taking care to rinse the probes with distilled water between each measurement to avoid cross-contamination.

The NOAA ARL HYSPLIT model (the National Oceanic & Atmospheric Administration and Air Resources Laboratory Hybrid Single-Particle Lagrangian Integrated Trajectory model) was used to run air trajectories using

Sample Period (18min)	Riming Level	CCN concentration (#/cc)	Number Mass Diameter (μm)	Snow Conductivity (amps/volt)	Snow pH Level	Collected Snow Mass (grams)	Rime Ice pH Level
1	1	306.9	10	4.9	5.2	4.55	3.89
2	3	47.7	15.6	7.4	5.12	6.25	4.27
3	5	23	21.6	9.65	5.05	12.89	4.52

Table 1. Table showing all values during sample periods

archived data from the week of March 11-18, 2006, to find possible origins of the air during the sample collection periods. The HYSPLIT model* was run using data from Grounded Data Analysis Software (GDAS). Trajectories were run backwards in time for 120 hours at three different heights above ground level. Knowing the origins of the air moving into the SPL region allows for assessment of whether the CCN levels in the cloud were affected by a polluted or non-polluted air mass.

RESULTS

The first sampling period took place on March 12, 2006 at 2245 UTC. The CCN concentration is 306.9 #/cc, while the average droplet diameter is 10 μm . The riming level is one. The snow conductivity is 4.9 amps/volts, and the snow pH level is 5.2. The overall snow mass collected is 4.55 grams. See Table 1 for a chart of values for all sampling periods. The calculated air trajectory during this sample period shows that the air passed through Northern Mexico and directly over Tucson, Arizona before arriving at SPL, indicating the air parcel is probably highly polluted. Mexican environmental laws are less robust than even

the United States, thus emissions from factories in Northern Mexico are not stringently regulated. The air in much of Northern Mexico is highly polluted (EIA, 2004). Tucson, AZ is also a city known for high amounts of air pollution (Eastwood, 2006). Also, no precipitation occurs before the air reaches SPL, thus no precipitation scavenging is occurring to remove pollutants from the air parcel (see Fig 2. A.).

The second sampling period was on March 15 at 2125 UTC. The CCN concentration is 47.7 #/cc. The average droplet diameter is 15.6 μm . Riming was assessed at a level of three. The snow conductivity is 7.4 amps/volts, and the snow pH level is 5.12. Snow mass collected was measure at 6.25 grams. The air trajectory for this exact time period is not shown, but is nearly identical to the trajectory shown in Fig 2. B. The air moves over California, Salt Lake City, UT, and again Tucson, AZ. The difference between this trajectory and the first trajectory is the precipitation values along the parcel path. A significant amount of precipitation occurs in excess of three hours just before the parcel reaches SPL. Considerable precipitation scavenging is indicated, thus many pollutants are rained out of the cloud before it reaches SPL.

The third sampling period occurred at 2231 UTC on March 5, 2006. The CCN concentration is 23.0 #/cc, and the average droplet diameter is 21.6 μm . Riming Level is five. Snow conductivity is 9.65 amps/volts. Snow pH level is 5.05. The snow mass collected is 12.89 grams. Fig. 2.

* A detailed explanation of the NOAA ARL HYSPLIT model can be found at

http://www.arl.noaa.gov/ready/hysp_info.html

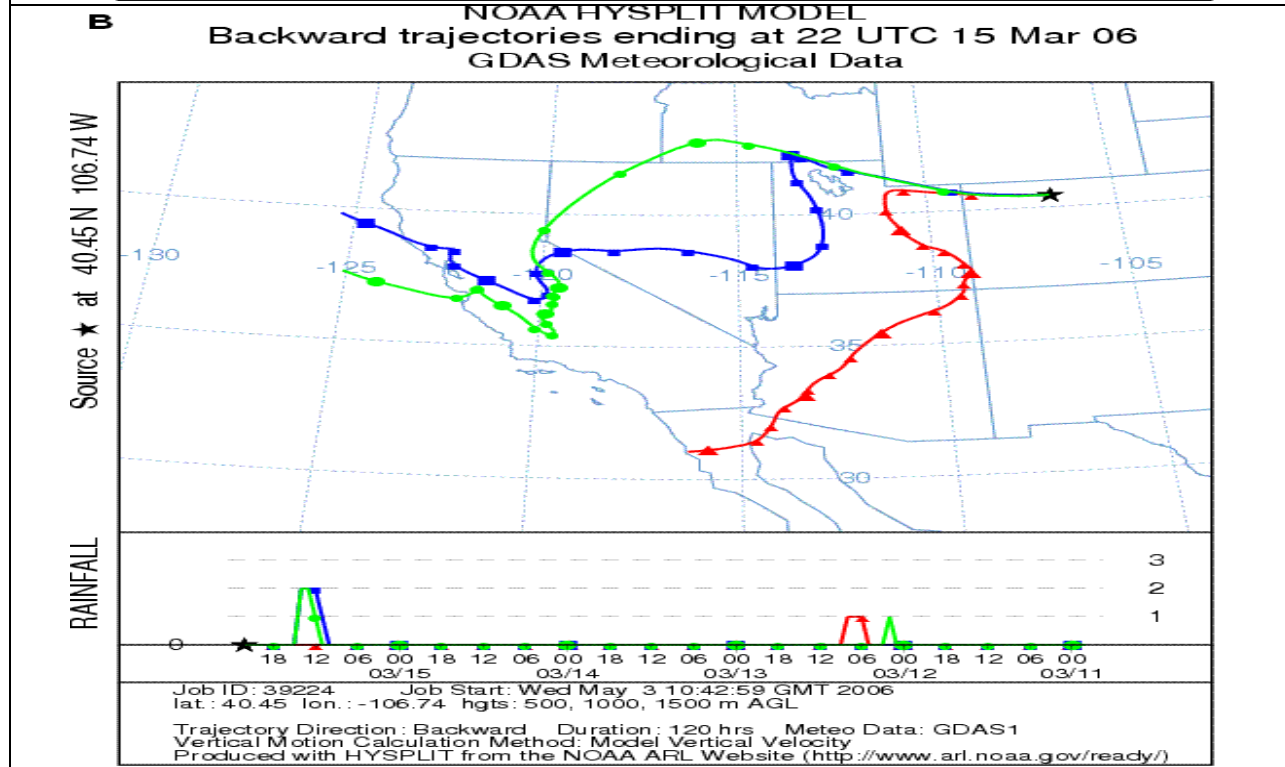
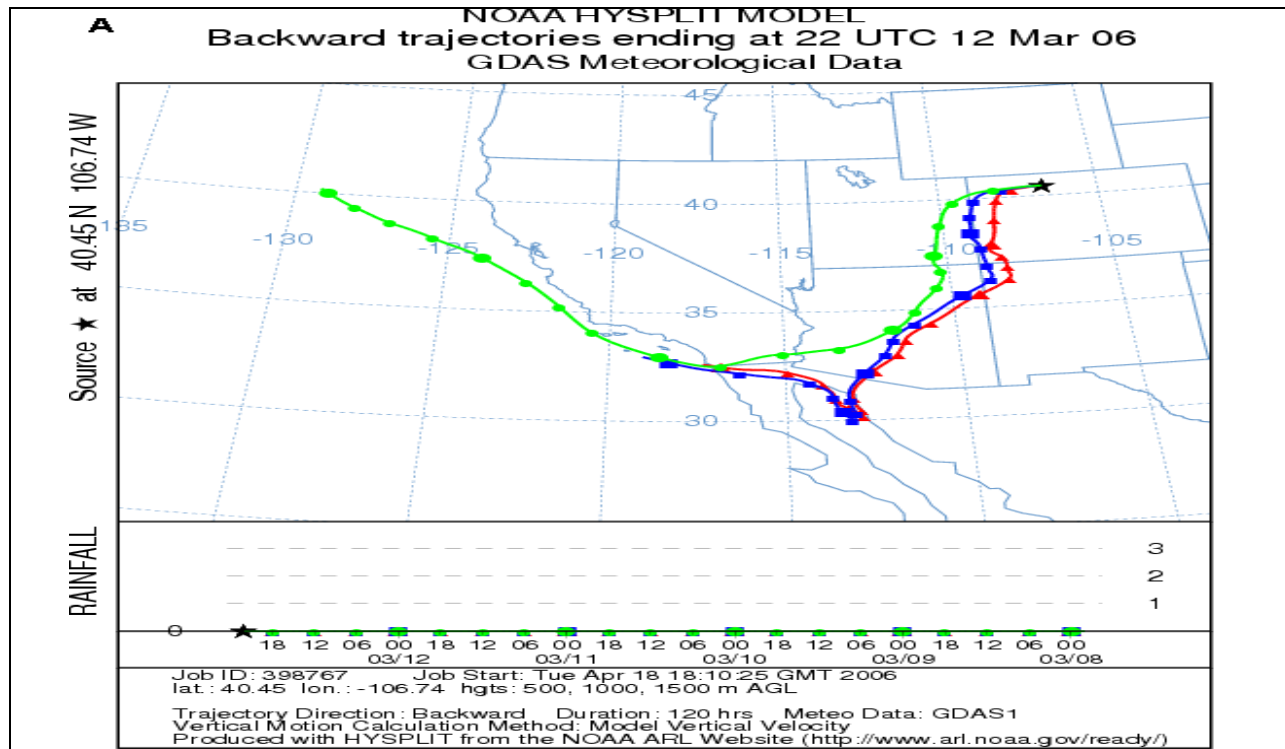


Fig. 2. A.) Backwards air trajectory calculated for 120 hours, and valid at 22 UTC on March 12, 2006 ending at SPL. The trajectory indicates high levels of pollution.
B.) Backwards air trajectory calculated for 120 hours, and valid at 22 UTC on March 15, 2006 ending at SPL. The trajectory indicates low levels of pollution.

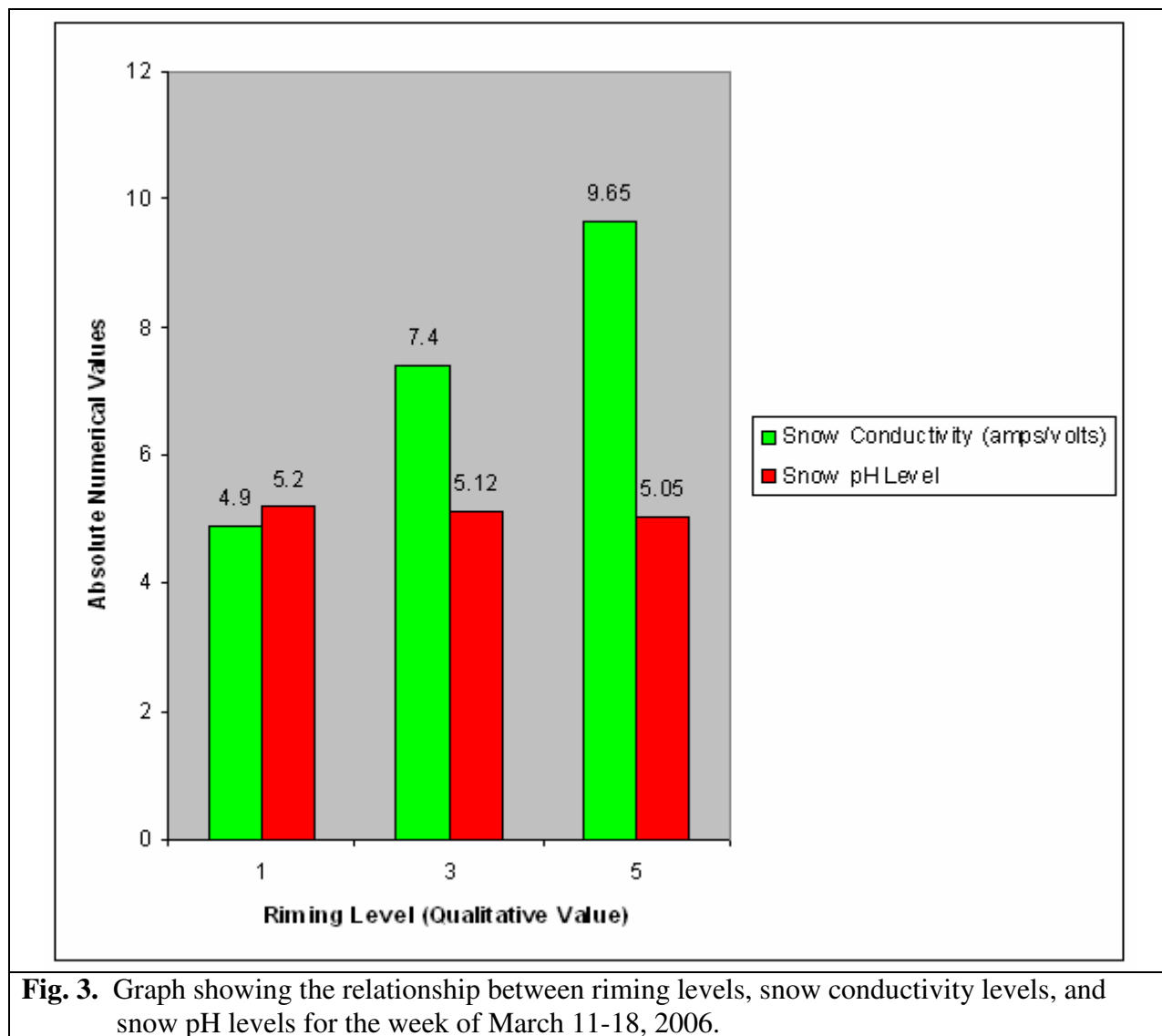


Fig. 3. Graph showing the relationship between riming levels, snow conductivity levels, and snow pH levels for the week of March 11-18, 2006.

B. shows the trajectory calculated for this time period. The pollution level in the air at this time is likely to be quite low, due to precipitation scavenging, as was explained in sampling period two.

DISCUSSION

The results of this study directly correspond to the findings of several previous studies conducted by other authors. Berg et al. (1990) concluded that high conductivity values but low pH values in snow samples can indicate high levels of riming. Also, according to Berg et al.

(1991), the chemical concentration of rime (acidity) is much higher than that of the chemical concentration of snow. When riming levels decrease, snow acidity should also decrease. Fig. 3 displays the relationship between riming levels, snow conductivity levels, and snow pH levels. As the riming levels increase, one sees a marked increase in snow conductivity, and a slight decrease in the acidity of the snow. This again shows that riming levels may be affected by features that change the conductivity and pH values of cloud droplets, such as atmospheric pollution. Fig 2. A. and Fig 2. B. indicate that pollution

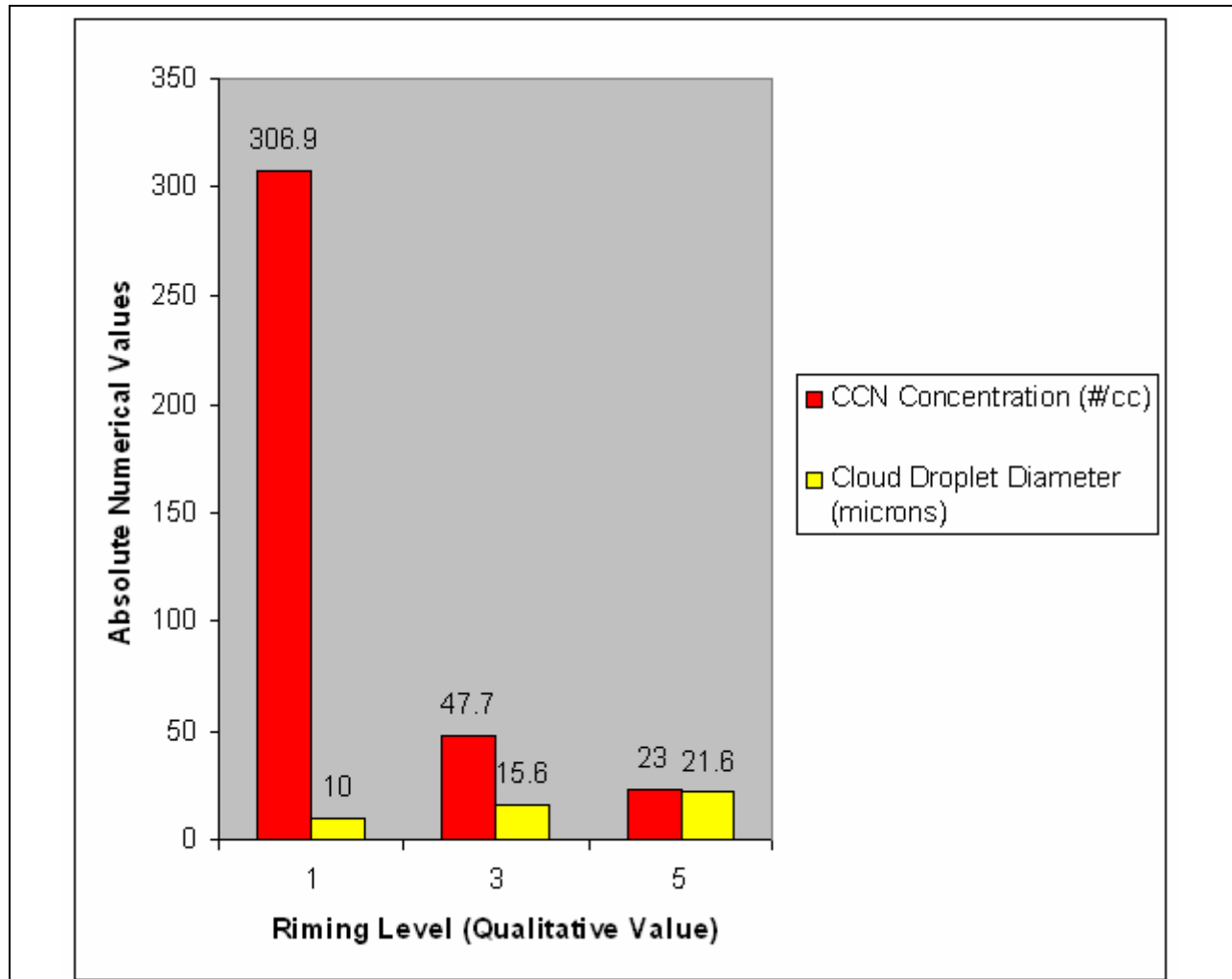


Fig. 4. Graph showing the relationship between Riming Level, CCN Concentration, and Cloud Droplet Diameter for the week of March 11-18, 2006.

levels are decreasing as the riming rate increases, thus pollution may have an adverse effect on riming levels.

Current research looks at the second indirect aerosol effect. This theory states that as CCN concentrations increase, the average diameter of cloud droplets within a cloud tend to decrease as the number of droplets within the cloud increase. Borys et al. (1999, 2003) proposed and showed that the second indirect aerosol effect also affects riming levels. Riming decreases (increases) as the number of cloud droplets increases (decreases), and the average cloud droplet diameter decreases (increases). Riming

decreases because droplets can become too small to rime onto crystals as CCN concentrations increase. If the droplets are too small, they may be swept around large falling ice crystals rather than riming directly onto the ice crystal. Fig. 4 shows the decreasing CCN concentrations correlated with an increase in droplet diameter and riming level, while Fig. 5 shows how riming levels, average droplet diameter, and total snow mass collected are affected by the changing CCN Concentrations. There is an inverse relationship between the CCN concentrations and the riming level, the

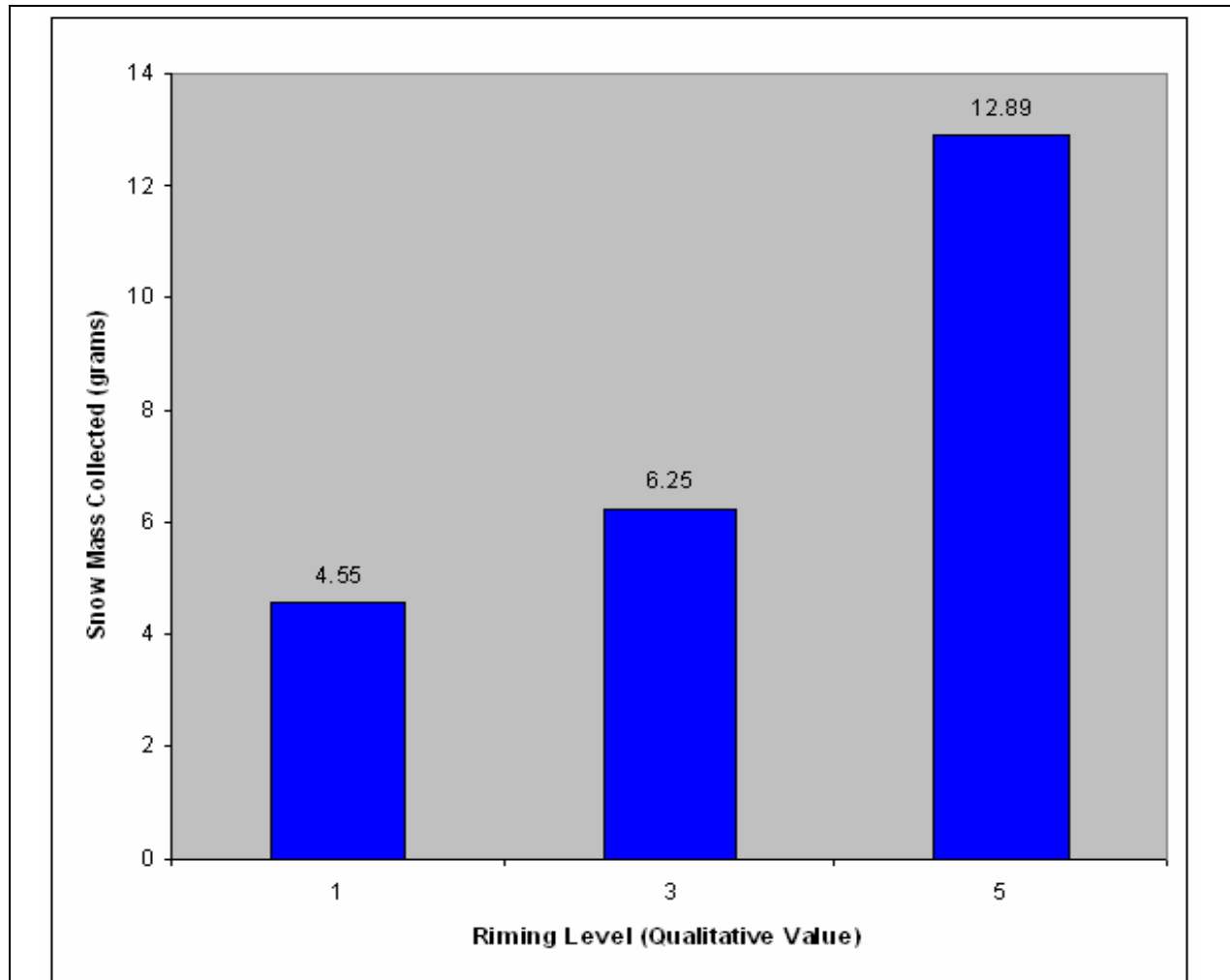


Fig. 5. Graph showing the relationship between Riming Level and Total Snow Mass Collected for the week of March 11-18, 2006

average cloud droplet diameter, and the total snow mass collected. As the average droplet diameter decreases due to increasing CCN concentrations, riming decreases, and thus the total mass of the snow decreases because less cloud water is freezing onto snow crystals. This mass decrease may be attributed to a decrease in precipitation rates during the sample period, or possibly a decrease in the snow water equivalent of the collected snow. The relationship is unclear based on this research.

The presented research supports the second indirect aerosol effect showing that CCN concentration affects riming levels. As CCN concentration increases, average

cloud droplet diameter and riming levels decrease. The data also shows a change in snow mass collected; the snow mass collected decreases as riming decreases, but the cause of this change is unknown. The change in snow mass may be caused by a change in the snow water equivalent of the collected snow, or possibly a change in the rate of precipitation. The effect of atmospheric aerosols on riming processes and subsequent precipitation quantities has recently become an issue of debate. If a change in riming level can affect precipitation rates, the climatology of the Western United States may be greatly influenced. Water is a highly lucrative

resource in this region, as water is scarce. The effect of pollution on riming and precipitation becomes a very significant issue.

In the most current research regarding the relationship between riming levels and precipitation, the possible correlation is analyzed from two different perspectives. Borys et al. (2003) performed a research study in the field, finding that a decrease in riming efficiency will decrease precipitation totals. The following year, an article was published by Lohmann. Lohmann used a GCM model to evaluate how decreased riming efficiency would affect global precipitation values, and found that precipitation totals would increase (2004).

Borys et al. (2003) addresses this issue with a study conducted in 2001 at the Storm Peak Laboratory in the northern Rocky Mountains of Colorado. "Mountaintop and remotely sensed data on aerosols and cloud and precipitation microphysics and chemistry were collected in wintertime synoptic scale storms systems to investigate the second indirect aerosol effect on precipitation formation" (Borys et al., 2003). This effect leads to reduced riming efficiencies, and according to Borys, a possible decrease in precipitation formation.

Measurements were recorded during two separate storm events. The first event was a storm in which very little riming occurred (shallow clouds with growth induced by diffusion), while the second event is a storm in which significant riming occurred (deep clouds with growth promoted by riming). Snow water equivalents (SWE's) were measured, and samples were taken at different times in each event, in order to analyze the cloud droplets and size of the snow crystals, the extent of the riming on the snow crystals, and the values of the snowfall rates. The snow was also analyzed for the anthropogenic sulfate concentrations, and these values were compared between the

two events in order to assess whether or not the clouds developed in a polluted air mass.

The results of this study show that on the unrimed day, concentrations of anthropogenic sulfate were high, concentrations of cloud droplets within the clouds were also high, and the average diameter of the cloud droplets was only 8.3 μm . On the rimed day, concentrations of anthropogenic sulfate were very low, concentrations of cloud droplets was over 2/3 less than on the unrimed day, and the average diameter of the cloud droplets was 13.6 μm . Snowfall rates and SWE's were much greater during the event which involved riming. In summation, on the rimed case day, the SPL experienced a "seven-fold higher precipitation rate" than in the unrimed case (Borys et al., 2003). The authors have thus concluded that a decrease in riming due to an increase in pollution, can lead to lower precipitation amounts.

Lohmann (2004) contradicts the findings of Borys et al. (2003). This article looks at "the climatic implications of ... (the Borys Study)... in a global climate model (GCM) simulation by replacing the constant riming efficiency with a size-dependent one appropriate for planar crystals and aggregates, respectively" (Lohmann, 2004). Lohmann states that the riming process depends on the shape of the crystal, and this detail was neglected in Borys et al. (2003). When this parameter is included, the "riming and the snowfall rate are initially increased in the polluted cloud due to its more numerous cloud droplets than in the clean cloud" (Lohmann, 2004). Precipitation amounts then depend on the type of snow crystal. If aggregate snow crystals are assumed, then the snowfall rate is actually increased by 40 percent, but if planar crystals are assumed, then the snowfall rates decrease by 30 percent. Lohmann used the ECHAM4 GCM using numerous variables and parameterizations to

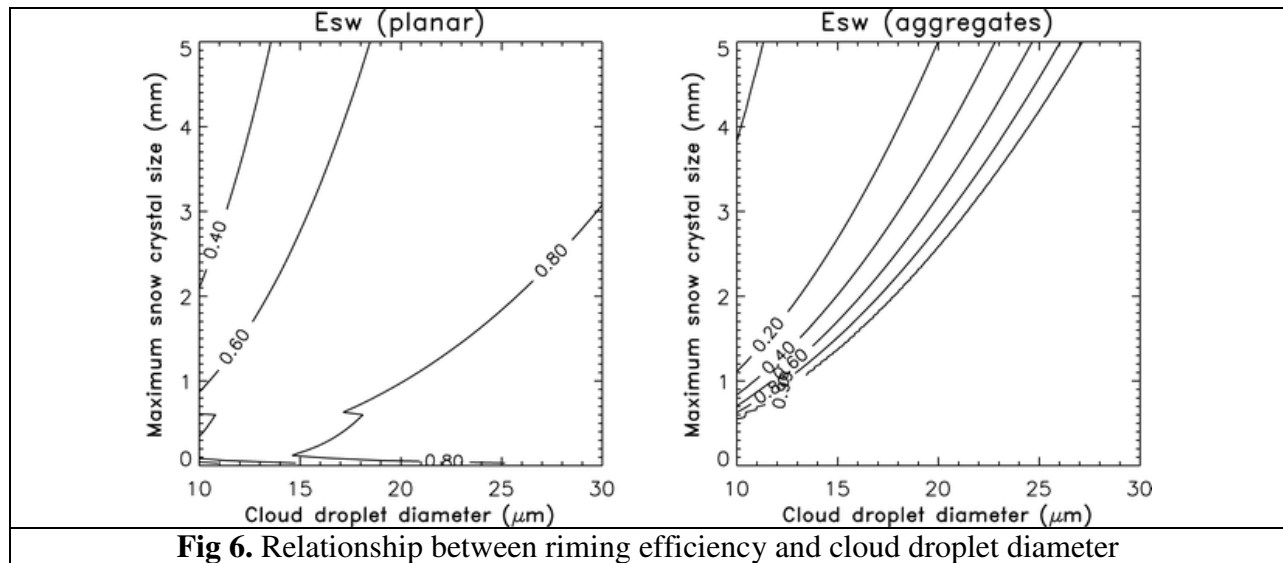


Fig 6. Relationship between riming efficiency and cloud droplet diameter

test this new hypothesis. Several equations were created to determine the riming efficiency of the two types of crystals as a function of droplet diameter and maximum crystal dimension, which is seen in Fig 6.

This figure shows that the riming efficiency “increases with cloud droplet diameter but decreases with snow crystal size” (Lohmann, 2004). For aggregates, the riming efficiency increases as the cloud droplet diameter gets larger than 11 μm , and the maximum size of the snow crystal increases. For planar crystals, the efficiency is between 0.4 and 0.8 for a large range of droplet and crystal sizes. When the size difference between snow crystals and cloud droplets increases, the efficiencies for the aggregate crystals span a larger range than that of the planar crystals.

Within the context of the GCM, Lohmann proposes that a,

“decrease in riming rate is...not translated in a reduced snowfall rate because the decrease in cloud droplet size, first of all, reduces the amount of solar radiation reaching the surface and thus cools the surface temperature over land and the atmospheric temperature in the Northern Hemisphere midlatitudes. This in turn favors depositional and

aggregational growth of ice and snow crystals thereby slightly increasing the snowfall rate in all present-day simulations over preindustrial times” (Lohmann, 2004).

Lohmann also proposes that Borys possibly recorded an isolated case in which the lowered riming rates reduced the snowfall rates, but this outcome is not representative of true nature on a global scale.

CONCLUSION

This research project corroborates the previous findings of Berg et al. (1990) and Borys et al. (2003). Namely, the analyzed data supports the second indirect aerosol effect, which states that an increase in CCN concentration will lead to an increase in cloud droplets, but a decrease in the average diameter of those droplets. The decrease in droplet diameter then affects riming within a cloud. As cloud droplet size decreases, the riming rates also decrease. The validation of previous studies provides a solid base upon which new conclusions may be drawn, such as correlations in the relationship between changes in riming and variations in precipitation rates. This current issue related to riming has reached an impasse.

Borys compared field observations during two separate precipitation events. The field observations indicate that a decrease in riming would also imply a decrease in the snowfall rates. Lohmann opposes this idea. The questions posed by Lohmann in the 2004 journal article, point out flaws in the Borys' et al. (2003) study because the research neglects the fact that the shape of the snow crystals, and not just the size and number of snow crystals, may affect the riming efficiency within a cloud. Lohmann's model run shows that a decrease in riming does not necessarily point to a decrease in the global snowfall rates. Snowfall rates may actually be increased as aggregate snow crystals form. While Lohmann proposes several interesting points, no field observations have been performed that can corroborate the findings of the GCM used. Two entirely different types of information are being compared. Model information is being used to deny actual field observations without anything tangible to support the theory. Borys et al. (2003) has actual numbers and measurements regarding riming and snowfall rates, while Lohmann is making a model-supported educated guess. Despite the fact that Borys et al. can support his claims with field measurements and data, his research team only analyzed two different snow events in order to draw conclusions. Further research is necessary to support the claims made by both of the research teams. More snow events should be examined and analyzed to be sure that conclusions drawn by Borys et al. (2003) are not just random and isolated events. The crystal structure of the snow should also be considered when evaluating whether the precipitation totals are increasing or decreasing, because Lohmann (2004) claims that this could completely change the outcome. Past research has looked at many different topics regarding riming including how chemicals

in the atmosphere affect the acidity of the snow, and how riming helps to retain certain trace gases within clouds and precipitation. This is related to the question concerning how pollutants are affecting atmospheric processes and changes in the global climate. Every day people around the world release more and more chemicals into the atmosphere without considering this very question. Hopefully, the research of scientists around the world will eventually make people reconsider their actions, and allow humans to make better decisions regarding the future regarding precipitation and water in the Western United States, and ultimately the future of this planet.

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